

# Quasi-phase-matching of high harmonic generation using polarization beating in optical waveguides

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A new scheme for quasi-phase matching high-harmonic generation is proposed in which polarization beating within a hollow core birefringent waveguide modulates the generation of harmonics. The evolution of the polarization of a laser pulse propagating in a birefringent waveguide is calculated and is shown to periodically modulate the harmonic generation process. The optimum conditions for achieving quasi-phase-matching using this scheme are explored and the growth of the harmonic intensity as a function of experimental parameters are investigated.

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## I. INTRODUCTION

High harmonic generation (HHG) is a nonlinear process in which odd multiples of a fundamental driving field are produced when an intense laser pulse is focused into a low density gas. HHG is an attractive source of temporally and spatially coherent, tuneable light with wavelengths in the XUV and soft X-ray range and has found applications in areas such as time resolved measurements [1–3], ultrafast holography [4], or diffractive imaging [5]. A semi-classical theory of HHG has been developed by Corkum [6] and a quantum treatment has been given by Lewenstein et al [7].

Although HHG is an attractive source for a wide range of experiments, its adoption in many applications is prevented by low conversion efficiency, resulting in low signal strengths. Typical conversion efficiencies for HHG are approximately  $10^{-6}$  at photon energies around 100eV and  $10^{-15}$  for photon energies above 1keV. This low conversion efficiency is caused by a phase mismatch between the driving laser field and the harmonics generated at each point in the generating medium. This results in the oscillation of the harmonic intensity with propagation distance, with an amplitude of 100%, preventing the continuous growth of the harmonic field. The period of this oscillation is  $2L_c$ , where  $L_c = \pi/\Delta k$  is the coherence length, and  $\Delta k$  is the phase mismatch. Phase mismatch arises from neutral gas and plasma dispersion and, if one is employed, the waveguide used to guide the driving radiation [8, 9].

When generating harmonics in a hollow-core waveguide it is possible to balance the dispersion due to the waveguide and free electrons with that of the neutral atoms. In this case true phase-matching,  $\Delta k = 0$ , can be achieved resulting in quadratic growth of the harmonic signal over extended regions. However, this approach is

limited to low levels of ionization, and consequently low laser intensities, due to the dominance of the free electron dispersion at higher ionization levels, placing a limit on the maximum harmonic order which can be phase-matched using this technique [8, 9]. Other phase matching schemes include difference frequency mixing or angular tuning of crossing beams have been proposed [10, 11]

An alternative approach to overcoming the phase mismatch is to suppress HHG in out-of-phase regions, a technique known as quasi-phase-matching (QPM). Various schemes have been developed for quasi-phase matching HHG. A series of gas jets, appropriately-spaced, has been used to achieve QPM [12, 13], but with this technique the total number of zones is limited by the Rayleigh range of the focused driving laser. Most quasi-phase matching schemes rely on guiding the driving laser pulse in a hollow-core waveguide since this extends the region over which harmonics can be generated. Experiments using a train of counter-propagating pulses that represses or scrambles the harmonic generation in the destructive zones have demonstrated quasi-phase-matching over up to 5 zones of harmonics with photon energies in the range [14–18]. Other QPM schemes include using a corrugated waveguide [19] or multi-mode beating [20, 21] in hollow-core waveguides.

In this paper we propose a new QPM technique – polarization-beating QPM (PBQPM) – which utilizes polarization beating in a birefringent waveguide to modulate the generation of harmonics [22]. The key advantage of PBQPM is its simplicity, since it avoids the need for additional laser pulses or longitudinally-structured gas targets. Instead, the only requirement is a birefringent waveguide with a suitable value of the birefringence.

In Section II the concept of PBQPM is outlined. Section III develops the theory of PBQPM, and Section IV explores the optimal parameters of this scheme. In Section V, birefringent waveguides and advantages of and limits to PBQPM are discussed.

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## II. POLARIZATION BEATING QPM

It is well known that the single-atom efficiency of HHG depends sensitively on the polarization of the driving laser field [23–26], which arises from the fact that the ionized electron must return to the parent ion in order to emit a harmonic photon.

In PBQPM a birefringent waveguide is used to generate beating of the polarization state of a driving linearly-polarized driving laser pulse, thereby modulating the harmonic generation process. QPM will occur if the period of polarization beating is suitably matched to the coherence length of the harmonics. In a birefringent waveguide, the incident radiation can be resolved into two components polarized along the birefringent axes. For linearly-polarized incident light these components are initially in phase, but the difference in their phase velocity will cause the two components to develop a phase difference which increases linearly with propagation distance. As a consequence, the resultant polarization state will evolve from linearly polarized at an angle  $\Theta$  to (for example) the fast axis, through (say) right-handed elliptical polarization to linearly polarized at an angle  $-\Theta$  to the fast axis, and thence through elliptical polarization back to linearly-polarized radiation parallel to the incident light.

The polarization beat length  $L_b$  is defined to be the distance for the two polarization components to develop a phase difference of  $\pi$ , and hence:

$$L_b \equiv \frac{\pi}{\Delta\beta} = \frac{\pi}{kB} = \frac{\lambda}{2B} \quad (1)$$

where  $\Delta\beta$  is the difference in propagation constant for the two polarization components,  $\lambda$  is the vacuum wavelength, and  $B = \frac{\lambda\Delta\beta}{2\pi}$  is the dimensionless birefringence parameter

Thus, by matching the beat length to an appropriate multiple of  $L_c$ , harmonic generation can be turned on and off along the length of the waveguide in a controlled way – enabling quasi-phase matching. Since the harmonics are generated most efficiently for linear polarization, the general condition for lowest-order for PBQPM is that the separation,  $L_b$ , of adjacent points of efficient harmonic generation is equal to an even number of coherence lengths, i.e. PBQPM requires,  $L_b = nL_c$ , where  $n$  is an even integer. As is discussed in more detail below, since the coherence lengths for harmonics polarized parallel to the  $\hat{x}$ - and  $\hat{y}$ -axes are different, for a given  $L_b$  it is only possible to quasi-phase-match one of these components. As a consequence the output harmonics will be predominantly linearly polarised along the matched birefringent axis. Moreover, as discussed below,  $\Theta$  and the harmonic order  $q$  determine the width of the generation zone in relation to  $L_b$  where  $\Theta$  can be optimized.

## III. THE ENVELOPE FUNCTION FOR PBQPM

If we write the electric field of the  $q$ th harmonic as

$$E_q(z, t) = \xi(z, t)e^{i[k(q\omega)z - q\omega t]} \quad (2)$$

then, within the slowly-varying envelope approximation:

$$\frac{\partial \xi}{\partial z} = A\Lambda(z)e^{-i[k(q\omega) - qk(\omega)]z} \quad (3)$$

where  $\xi$  is the electric field envelope,  $k(\omega) = \beta(\omega)$  is the propagation constant for radiation of frequency  $\omega$ ,  $A$  is a normalization constant and  $\Lambda(z)$  is a relative source term.

For each polarization the wave vector mismatch for a specific harmonic,  $q$ , can be written as:

$$\Delta k = k(q\omega) - qk(\omega) \quad (4)$$

$$= \Delta k_{\text{plasma}} + \Delta k_{\text{neutral}} + \Delta k_{\text{waveguide}} \quad (5)$$

and the envelope function can be solved as:

$$\xi(z) = A \int_0^z dz' e^{-i\Delta k z'} \Lambda(z'). \quad (6)$$

### A. Effect of polarization state on HHG

The relative number of harmonic photons generated in the  $q$ th harmonic by a driving beam of ellipticity  $\varepsilon$  may be written as:

$$f(\varepsilon) \approx \left( \frac{1 - \varepsilon^2}{1 + \varepsilon^2} \right)^\alpha \quad (7)$$

where  $\varepsilon$  is defined as the ratio between the minor axis to major axis.

Within the perturbative regime  $\alpha = q - 1$ , as verified by Budil et al [23] for harmonics  $q = 11$  to 19, and by Dietrich et al for harmonics up to  $q \approx 31$  [27]. Schulze et al found that for higher-order harmonics the sensitivity of harmonic generation to the ellipticity of the driving radiation is lower than predicted by Eqn (7) with  $\alpha = q - 1$  [28], although in this non-perturbative regime the efficiency of harmonic generation still decreases strongly with  $\varepsilon$ . Further measurements of the dependence of harmonic generation on ellipticity have been provided by Sola et al [24]. It is recognized that Eqn (7) is an approximation, but it will serve our purpose of demonstrating the operation of PBQPM.

The offset angle and ellipticity of the harmonics generated by elliptically-polarized radiation have been shown to depend on the ellipticity and intensity of the driving radiation, and on the harmonic order [25, 26, 28–30]. Propagation effects can also play an important role. Since the amplitude with which harmonics are generated decreases strongly with increasing ellipticity, we are most interested in the ellipticity of the harmonics generated for

small  $\varepsilon$ . It has been shown that for higher-order harmonics, and/or high driving intensities, both the ellipticity and change in ellipse orientation of the harmonics generated by radiation with  $\varepsilon \approx 0$  are close to zero [25]. We will therefore make the simplification that the generated harmonics are linearly polarized along the major axis of the driving radiation, and that the harmonics polarized along the fast and slow axes of the waveguide may be treated separately.

### B. Birefringence & evolution of the ellipticity of polarization

In this section, the evolution of the driving field's ellipticity will be developed. We assume azimuthal symmetry of the modes. Let  $\hat{x}$  and  $\hat{y}$  be the birefringent axes of the waveguide with the  $\hat{x}$  axis being the slower axis such that  $\beta_x < \beta_y$ . The driving field can be decomposed into the  $\hat{x}$  and  $\hat{y}$  components:

$$\vec{\mathcal{E}}(r, z, t) = \begin{pmatrix} \mathcal{E}_x \\ \mathcal{E}_y \end{pmatrix} = E(r) \begin{pmatrix} e^{i[(\beta_y - \Delta\beta)z - \omega t]} \sin \Theta \\ e^{i[(\beta_y)z - \omega t]} \cos \Theta \end{pmatrix} \quad (8)$$

where  $E(r)$  describes the transverse electric field profile as a function of the distance  $r$  from the propagation axis.

At a given point  $z$  the electric field has an ellipticity given by:

$$\varepsilon(z) = \sqrt{\frac{\sin^2 \Theta \cos^2(\phi_{min} - \Delta\beta z) + \cos^2 \Theta \cos^2(\phi_{min})}{\sin^2 \Theta \cos^2(\phi_{max} - \Delta\beta z) + \cos^2 \Theta \cos^2(\phi_{max})}} \quad (9)$$

where

$$\tan^2 \Theta = -\frac{\sin[2\phi]}{\sin[2(\phi + \Delta\beta z)]} \quad (10)$$

Here  $\phi_{max}$  and  $\phi_{min}$  are the values of  $\phi = \omega t$  which give respectively the maximum and minimum magnitude of the electric field.

Although the ellipticity is periodic with period  $L_b$ , the electric field is periodic with period  $2L_b$ . The beating of the ellipticity as a function of propagation distance for various different angles of incidence is shown in Fig. 1.

### C. Wave-vector mismatch in a birefringent waveguide

In a birefringent waveguide the wave vector mismatch  $\Delta k$  is polarization-dependent. However, the only polarization-dependent term in Eqn (5) is  $\Delta k_{waveguide}$ , the wave vector mis-match arising from the waveguide dispersion. Hence we may write,

$$\begin{cases} \Delta k_{x, waveguide} = \beta_x(q\omega) - q\beta_x(\omega) \\ \Delta k_{y, waveguide} = \beta_y(q\omega) - q\beta_y(\omega) \end{cases} \quad (11)$$

From this, we may then write,

$$\Delta k_x = \Delta k_y + \delta k_{xy},$$

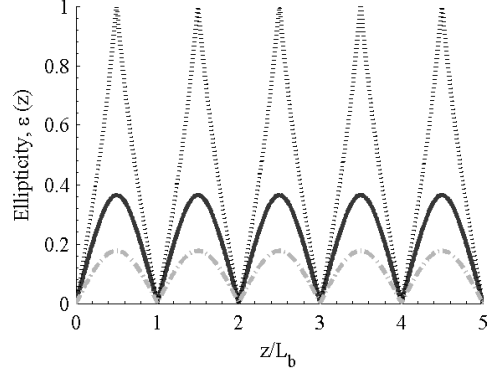


FIG. 1. Ellipticity beating for different incidence angles  $\Theta$ . From top to bottom: Dotted light gray line shows  $\Theta = 45^\circ$ , Solid gray line shows  $\Theta = 30^\circ$  and  $\Theta = 70^\circ$ ; and dot-dashed gray line shows  $\Theta = 20^\circ$  and  $\Theta = 80^\circ$ .

where,

$$\delta k_{xy} = \Delta k_{y, waveguide} - \Delta k_{x, waveguide} \quad (12)$$

$$= q\Delta\beta(\omega) - \Delta\beta(q\omega) \quad (13)$$

$$\approx q\Delta\beta(\omega) \quad (14)$$

and the approximation follows if the birefringence is small at the frequency of harmonic  $q$ .

### D. Constructing the PBQPM envelope function

From the discussion in Section III A we may treat each polarization separately, and hence Eqn (6) becomes:

$$\begin{cases} \xi_x(z) = A \int_0^z dz' e^{-i\Delta k_x z'} \Lambda_x(z') \\ \xi_y(z) = A \int_0^z dz' e^{-i\Delta k_y z'} \Lambda_y(z') \end{cases} \quad (15)$$

where

$$\begin{cases} \Lambda_x(z') = \sqrt{f[\varepsilon(z')]} \sin \Theta \\ \Lambda_y(z') = \sqrt{f[\varepsilon(z')]} \cos \Theta \end{cases} \quad (16)$$

are the relative source terms. In terms of the faster polarization state (the  $\hat{y}$  component),

$$\begin{cases} \xi_x(z) = A \sin \Theta \int_0^z dz' e^{-i(\Delta k_y + q\Delta\beta)z'} \sqrt{f[\varepsilon(z')]} \\ \xi_y(z) = A \cos \Theta \int_0^z dz' e^{-i\Delta k_y z'} \sqrt{f[\varepsilon(z')]} \end{cases} \quad (17)$$

Fig. 2 shows an example of  $f[\varepsilon(z)]$  and the real part of the envelope function integrand, assuming  $\alpha = q - 1$  and  $L_b = 2L_{c,y}$ . Notice that integrating across the  $\hat{y}$  component would result in a monotonic increase of the HHG amplitude whereas for the  $\hat{x}$  component, no net increase in harmonic amplitude would result owing to the rapid oscillations of the integrand. We conclude that the polarization of the output harmonic will be almost perfectly linear, in this case with an electric field parallel to the  $y$ -axis since it is for that polarization that the

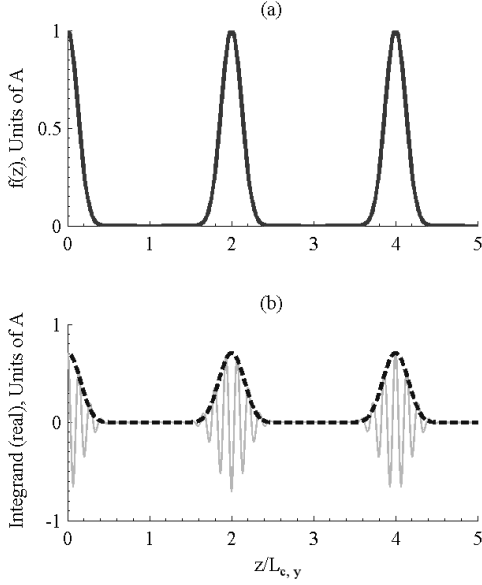


FIG. 2. Top graph (a): Variation of the harmonic generation efficiency,  $f(z) = f[\varepsilon(z)]$ , as function  $z$  for  $q = 27$  and  $\Theta = 45^\circ$ . Bottom graph (b): Real part of the of the  $\hat{x}$  component of the integrand (solid gray) and  $\hat{y}$  component (dashed black line). Assuming  $L_b = 2L_{c,y}$ ,  $\Theta = 45^\circ$  and  $q = 27$

polarization beating has been matched to the coherence length

Fig. 3 compares PBQPM for these parameters against perfect phase matching and perfect QPM; we define the latter to correspond to square-wave modulation of the local harmonic generation with a period  $2L_c$  and complete suppression of the out of phase zones.

#### IV. PROPERTIES OF PBQPM

##### A. Coherence length and beat length matching

For a waveguide with fixed birefringence  $B$  the condition for PBQPM can be realized by tuning the gas pressure and/or adjusting the driving laser intensity until the period of polarization beating and the coherence length are appropriately matched. The general condition for QPM is that the distance over which harmonics are generated and that over which generation is suppressed are both equal to an odd number of coherence lengths, i.e. to  $(2l+1)L_c$  and  $(2m+1)L_c$  respectively, where  $l$  and  $m$  are integers. Thus the QPM period is in general  $2(l+m+1)L_c = nL_c$  and hence the general condition for PBQPM is  $L_b = nL_c$ , where  $n$  is even. Note that if the distances over which harmonics are generated and suppressed are equal, i.e.  $l = m$ , then PBQPM requires satisfaction of the more restrictive condition  $L_b = 2(2l+1)L_c$ . These considerations are illustrated by Fig. 4 which shows the calculated growth of the

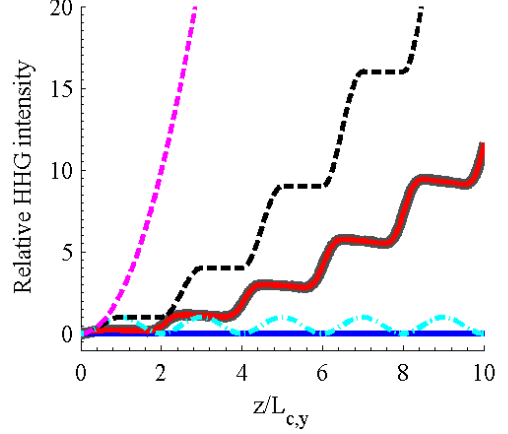


FIG. 3. Relative HHG intensity for different phase matching conditions assuming that  $L_b = 2L_{c,y}$ ,  $q = 27$ ,  $\Theta = 20^\circ$  for PBQPM. Dashed magenta line shows perfect phase matching, dashed black line for perfect quasi-phase matching, dot-dashed cyan line for no phase matching. The solid blue line shows the relative amplitude squared for the  $\hat{x}$  component whilst the dashed yellow line shows the relative amplitude squared for the  $\hat{y}$  component. The thick gray line shows the total HHG intensity for PBQPM.

harmonic intensity for the cases  $n = 1, 2, 3$  and 4. It may be seen that monotonic growth of the harmonics does not occur when  $n$  is odd, as expected. Notice that PBQPM does occur for the case  $n = 4$ , which corresponds to the lengths of the suppressed and unsuppressed regions of harmonic generation being unequal; this is possible for PBQPM since the efficiency of harmonic generation is very sensitive to the ellipticity, so that it is possible for the harmonic generation regions to be shorter than those in which generation is suppressed.

##### B. Output intensity and coupling angle $\Theta$

It can be expected that the coupling angle  $\Theta$  has an optimum: if  $\Theta$  is small then the  $\varepsilon$  remains small for all  $z$ , and the harmonic generation is not sufficiently suppressed in the out of phase zones; if it is too large then harmonics are only generated for a small fraction of the in-phase zones. Fig. 5 shows the relative harmonic intensity as a function of propagation distance for various coupling angles  $\Theta$ . Fig. 6a shows as a function of  $\Theta$ , and for various harmonics  $q$ , the normalized harmonic signal after one beat length. Also shown is the optimum coupling angle as a function of harmonic order  $q$ .

The optimum coupling angle,  $\hat{\Theta}$ , will decrease with harmonic order since higher-order harmonics are more sensitive to the ellipticity of the driving radiation as discussed in Section III B, and hence for these harmonics it is possible to couple more of the polarization of the incident radiation along the polarization of the harmonics to be generated by PBQPM. Fig. 6b confirms this for the

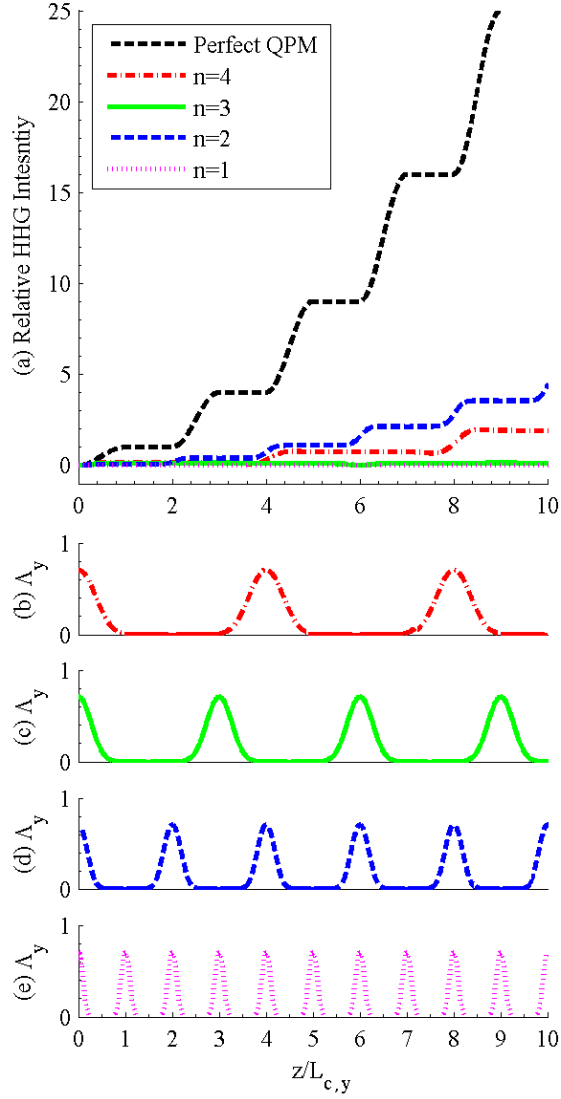


FIG. 4. PBQPM for  $\Theta = 34^\circ$ ,  $q = 27$ , and  $L_b = nL_{c,y}$  with  $n = 1, 2, 3, 4$ . The top graph shows the relative harmonic intensity as a function of propagation distance  $z$  for perfect QPM (dashed black line),  $L_b = 4L_{c,y}$  (dot-dashed red line),  $L_b = 3L_{c,y}$  (solid green line),  $L_b = 2L_{c,y}$  (dashed blue line), and  $L_b = L_{c,y}$  (dotted magenta line). The bottom four graphs show the  $p = y$  polarization source term  $\Delta_y$  as a function of propagation distance  $z$  for (b)  $L_b = 4L_{c,y}$ , (c)  $L_b = 3L_{c,y}$ , (d)  $L_b = 2L_{c,y}$ , and (e)  $L_b = L_{c,y}$ .

case of  $L_b = 2L_{c,y}$ .

## V. DISCUSSION

Having introduced the concept of PBQPM and explored its operation in theory, we now discuss how this

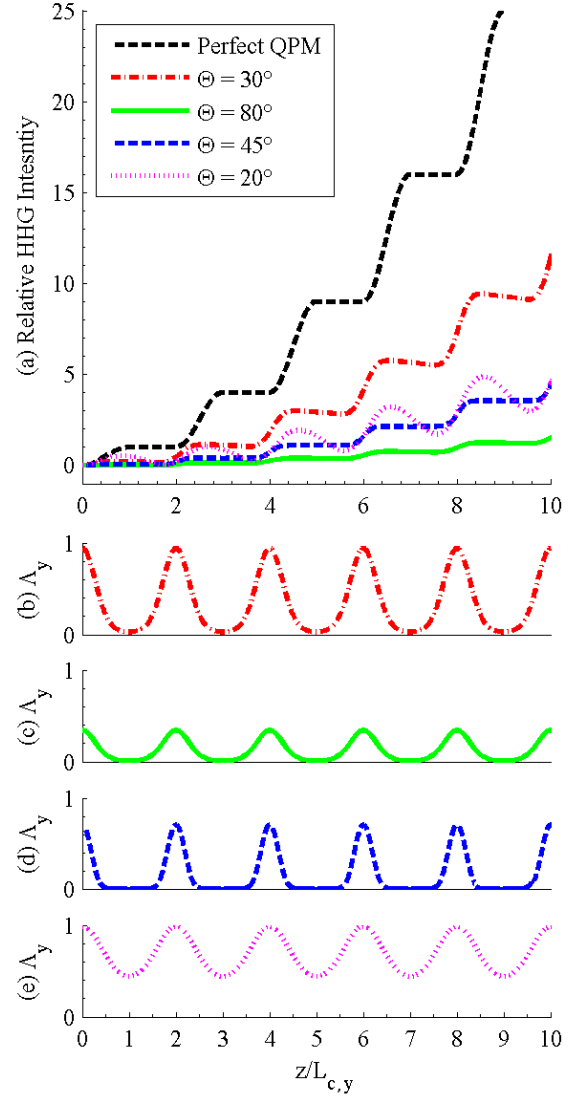


FIG. 5. PBQPM for  $q = 27$  and  $L_b = 2L_{c,y}$  with  $\Theta = 30^\circ, 80^\circ, 45^\circ, 20^\circ$ . The top graph shows the relative harmonic intensity as a function of  $z$  for perfect QPM (dashed black line) and coupling angles of  $\Theta = 30^\circ$  (dot-dashed red line),  $\Theta = 80^\circ$  (solid green line),  $\Theta = 45^\circ$  (dashed blue line),  $\Theta = 20^\circ$  (dotted magenta line). The bottom four groups show the  $p = y$  polarization source term  $\Delta_y$  as a function of  $z$  for (b)  $\Theta = 30^\circ$ , (c)  $\Theta = 80^\circ$ , (d)  $\Theta = 45^\circ$ , and (e)  $\Theta = 20^\circ$ .

quasi-phase-matching scheme might be realized in practice. Several types of birefringent waveguides have been developed to date: elliptical or rectangular waveguides [31]; waveguides made from birefringent materials [32]; waveguides with nonuniform refractive index [31]; photonic crystal fibers (PCFs) [33–35]; waveguides constructed with tunable piezoelectric material [36]; tunable stress-induced birefringent waveguides [31]. Many

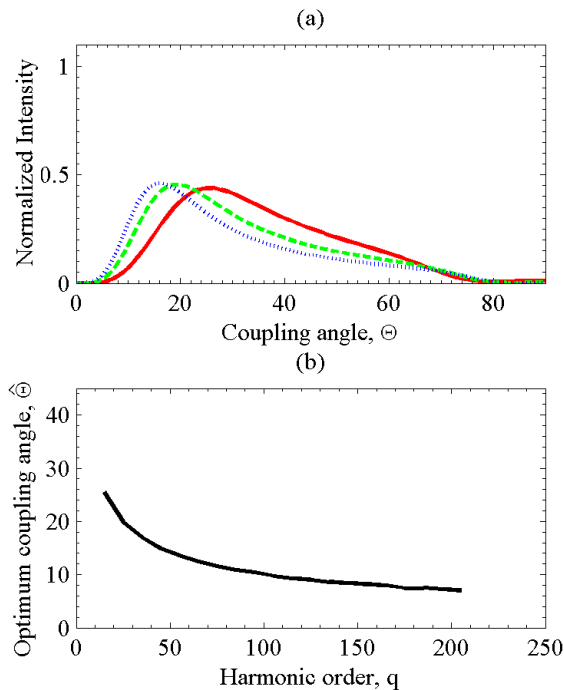


FIG. 6. Top graph (a): Relative HHG intensity as a function of coupling angle  $\Theta$ , normalized to perfect QPM after one beat length where  $L_b = 2L_{c,y}$  for  $q = 15$  (solid red line),  $q = 27$  (dashed green line),  $q = 39$  (dotted blue line). Bottom graph (b): Optimum angle  $\hat{\Theta}$  as a function of harmonic order  $q$  for  $L_b = 2L_{c,y}$

of these birefringent waveguides employ solid cores, and are therefore not suitable for PBQPM. However, we note that (non-birefringent) gas-filled PCFs have been used to generate high harmonics [37]. Moreover, commercially available hollow core birefringent PCFs have been demonstrated with birefringence parameters as large as  $B > 10^{-4}$  [35, 38], corresponding to matching a coherence length of  $\sim 1$ mm, and solid-core PCFs with  $B > 10^{-31}$  have been demonstrated [39]. The birefringence of optical crystals can be as large as  $B \approx 0.4$  [40]. Although the birefringence of hollow-core waveguides made from materials of this type would be only a fraction of that of the wall material, it seems likely that construction of hollow-core waveguides with sufficient birefringence to match coherence lengths in the range of a few hundred microns will be possible. The development and study of hollow-core birefringent waveguides of this type will form the basis of future work.

The principal advantage of PBQPM is the simplicity of the experimental set up: a single laser pulse correctly coupled into a birefringent waveguide. Other QPM techniques are likely to be limited by several factors: for pulse-train QPM, the number of ultrafast pulses which can be generated; the precision with which corrugated waveguides or gas jet arrays can be manufactured; or, for multi-mode QPM, different damping rates for the two

waveguide modes. PBQPM does not suffer from these difficulties. As for any HHG scheme limits will be set by absorption of the harmonics and variation of the ionization level – and hence of the coherence length – within the driving pulse; and as for other techniques employing a waveguide, damping of the driving radiation may, in principle, limit the length over which harmonics will be generated. A potential limitation unique to PBQPM is dispersion of the two polarization components: the slower polarization will lag the faster one until the two polarization states separate and polarization beating ceases. In principle this limitation could be overcome by reversing the birefringence after the two polarization components have slipped significantly; the limits imposed by all these mechanisms will be explored in future work.

Finally we note that it is also possible to achieve PBQPM in non-birefringent waveguides by exciting different-order waveguide modes with nonparallel polarizations. In this arrangement the different phase velocities of the modes will lead to polarization beating in addition to the beating of the intensity which is utilized in multi-mode QPM [20]. Indeed, driving PBQPM with different-order waveguide modes increases the available parameter space and could allow higher-order harmonics to be quasi-phase-matched and greater efficiencies to be achieved.

## VI. CONCLUSION

We have proposed a new method for quasi-phase-matching HHG – polarization-beating QPM (PBQPM) – in which polarization beating in a birefringent waveguide is employed to modulate the efficiency with which harmonics are generated. Quasi-phase-matching may then be achieved by suitably matching the period of polarization beating with the coherence length.

We have developed a theoretical model of PBQPM and used this to demonstrate that PBQPM can indeed enhance the intensity with which harmonics can be generated. This model was also used to explore the optimum coupling angle for PBQPM and to show that this depends on the harmonic order  $q$ .

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